

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
5 July 2001 (05.07.2001)

PCT

(10) International Publication Number
WO 01/48852 A1

(51) International Patent Classification⁷: H01M 8/04, 8/24

(21) International Application Number: PCT/US00/32542

(22) International Filing Date:
30 November 2000 (30.11.2000)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
09/472,388 23 December 1999 (23.12.1999) US

(71) Applicant: **THE REGENTS OF THE UNIVERSITY OF CALIFORNIA** [US/US]; Los Alamos National Laboratory, LC/BPL, MS D412, Los Alamos, NM 87545 (US).

(72) Inventors: **REN, Xiaoming**; 1340 44th Street, Los Alamos, NM 87544 (US). **GOTTESFELD, Shimshon**; 625 Paige Loop, Los Alamos, NM 87544 (US).

(74) Agents: **WILSON, Ray, G.** et al.; Los Alamos National Laboratory, LC/BPL, MS D412, Los Alamos, NM 87545 (US).

(81) Designated States (*national*): AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, UZ, VN, YU, ZW.

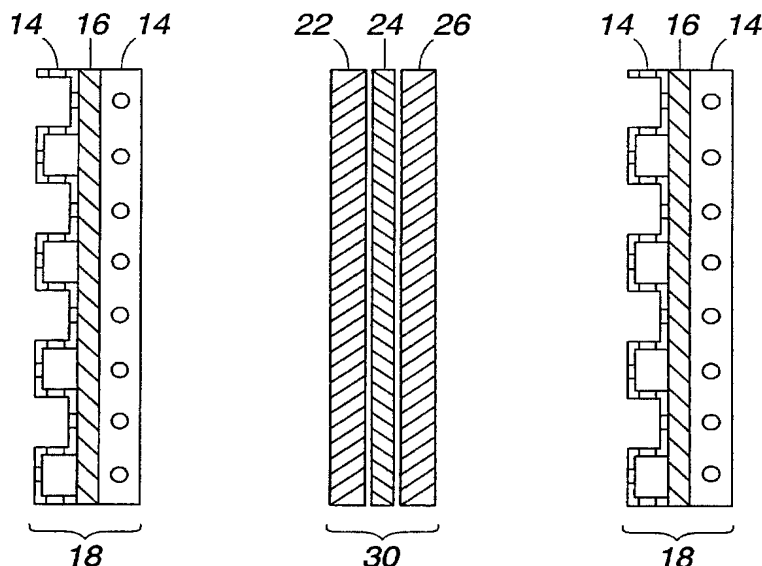
(84) Designated States (*regional*): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

Published:

— With international search report.

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: FLOW CHANNEL DEVICE FOR ELECTROCHEMICAL CELLS



(57) Abstract: A compact, inexpensive bipolar plate assembly (18) for use in electrochemical cells, such as fuel cell and electrolysis cells, is formed from a sandwich of metal plates. A smooth electrically conductive sheet (16) having first and second sides provides a sealing and reactant separator member. In one embodiment, first and second sheets (14) of corrugated material define flow channels on both sides, and have perforations in the material to permit fluid transfer between the two flow channels. This corrugated sheet are arranged on opposite sides of the smooth conductive sheet. Lands defined by the corrugations establish electrical contacts with a node (22) and cathode (26) portions of adjacent electrochemical cells (30). The cells can be further connected in series to form a stack that delivers a selected power output.



WO 01/48852 A1

FLOW CHANNEL DEVICE FOR ELECTROCHEMICAL CELLS

5 STATEMENT REGARDING FEDERAL RIGHTS

This invention was made with government support under Contract No. W-7405-ENG-36 awarded by the U.S. Department of Energy. The government has certain rights in the invention.

10 FIELD OF THE INVENTION

The present invention relates generally to flow channel plates for electrochemical cells, and, more particularly, to flow channel plates useful to form bipolar plates, coolant plates, and the like, for electrochemical cells.

15 BACKGROUND OF THE INVENTION

Flow channel plates, e.g., bipolar plates, coolant plates, and the like, for electrochemical cells, such as fuel cells, electrolysis cells, and the like, are typically formed from graphite or metal with machined flow channels or from various carbon composite materials with machined or molded flow channels. For applications to fuel
20 cell stacks, the flow field plates serve in bipolar plates with an anode flow field on one side of a plate and a cathode flow field on the opposite side of the plate. It has been difficult to achieve compact fuel cell stacks with such flow fields, particularly where low air flows are involved and whereas low pressure drop across the stacks must be obtained. For applications to electrochemical cells for electrolysis, the flow channel
25 plates provide for the delivery of liquid or gaseous reactants and removal of the electrolysis products.

U.S. Patent 5,482,792, issued January 9, 1996, teaches the use of porous deformable structures ("collectors") for flow fields to distribute gaseous reactants over membrane backings while permitting transverse flows to enhance uniform distribution
30 of the reactants. Separate structures are provided as bipolar plates. The porous structures are taught as metal-wire matrices or screens mounted in a support frame

structures are taught as metal-wire matrices or screens mounted in a support frame and do not provide any structural support. The fluid flow must traverse the matrix or screen structure since flow channels are not provided.

5 U.S. Patent 4,755,272, issued July 5, 1988, teaches the use of a corrugated, electrically conductive bipolar plate having separated flow fields for supplying fuel and oxidant on opposite sides of the corrugated plate, where the corrugations are slightly compressed between two electrodes held in a cell body.

10 Exemplary flow channel configurations for use in electrolysis cells are shown in U. S. Patent 4,191,618, issued March 4, 1980. As shown therein, simple flow channels are formed from parallel ribs with an overlying perforated plate serving as the anode.

15 Various objects, advantages and novel features of the invention will be set forth in part in the description which follows, and in part will become apparent to those skilled in the art upon examination of the following or may be learned by practice of the invention. The objects and advantages of the invention may be realized and attained by means of the instrumentalities and combinations particularly pointed out in the appended claims.

SUMMARY OF THE INVENTION

20 A compact, inexpensive flow distribution assembly for use in electrochemical cells, such as fuel cells, is formed from a sandwich of metal sheets. A smooth electrically conductive sheet having first and second sides provides a separating member between adjacent electrochemical cells. A first sheet of conductive material has corrugations that define flow channels and has perforations in the material to
25 permit fluid transfer between the alternating flow channels. In one embodiment, a second corrugated is provided on the second side of the conductive sheet, whereby the first corrugated sheet is arranged on the first side of the smooth conductive sheet and the second corrugated sheet is arranged on the second side of the smooth conductive sheet. Lands defined by the corrugations establish electrical contacts with
30 anode and cathode portions of adjacent electrochemical cells.

BRIEF DESCRIPTION OF THE DRAWINGS

The accompanying drawings, which are incorporated in and form a part of the specification, illustrate embodiments of the present invention and, together with the description, serve to explain the principles of the invention. In the drawings:

FIGURES 1A and 1B illustrate formation of a corrugated flow field device in accordance with one embodiment of the present invention.

FIGURE 2 is an exploded view of a flow field assembly according to one embodiment of the present invention.

FIGURE 3 is a cross-sectional view of an electrochemical cell using the flow channel assemblies shown in FIGURE 2.

FIGURE 4 is an isometric cross-section view of an electrochemical cell stack using electrochemical cells shown in FIGURE 3.

DETAILED DESCRIPTION

Electrochemical cells based on polymer electrolytes are frequently operated with a liquid flow across the anode of each unit cell and a liquid/gas flow at the cathode of each unit cell. The gas supplied to the cathode is often oxygen or air. Flow channel devices serve to direct the flows over backing/catalytic plates that contact opposed faces of an electrolyte membrane that conducts ions to complete an electrical circuit. Effective flow channel devices serve to uniformly distribute the appropriate flow over a face of the associated backing plate with a small pressure drop across the flow channel device.

The present invention provides a flow field device formed from a perforated, corrugated metal sheet. The corrugations provide flow channels and the perforations permit the reactant flow to redistribute between channels, particularly channels having a liquid and/or gas flow restriction, to maintain a uniform flow over the backing plates. By using such a flow field, stacks of cells with compact unit cell packing size can be achieved with minimum pressure drop across the cell.

In accordance with the present invention, a flow field device 14, shown in Figures 1A and 1B, is formed from a plate 10 having perforations 12. Perforations 12 are preferably in a staggered arrangement to maintain structural integrity of plate 10, but many variations of perforations can be provided.

5 To form a flow distribution device 14, perforated plate 10 is corrugated, as shown in Figure 1B, i.e., plate 10 is formed into a configuration having folds of alternating ridges and valleys, where the contour of the corrugations may be smoothly varying, e.g., sinusoidal, or be substantially square, e.g., with flat ridges and valleys, or triangular. The contact resistance between the ridges and adjacent conductive
10 surfaces will determine what configurations are acceptable in any given design. In a preferred embodiment, flow distribution device 14 is formed with substantially square corrugations having a spacing effective to place perforations in a manner that permits transverse flow distribution along corrugated plate 14 and longitudinal flow through the lands that form the ridges and valleys.

15 In an experimental, non-optimized embodiment, the perforations accounted for up to 50% of the area of sheet 10. To ensure mechanical integrity under compression in an operating cell and to minimize electrical contact resistance with adjacent cell components, the sheets were 4 mil thick stainless steel that was electrochemically plated with a 200 nm thick gold layer. Acceptable contact resistance is also obtained
20 from perforated sheets of stainless steel alloys of the 300 and 400 series where a high compression axial loading is applied to the cell. Other conductive metals may be found to be useful by routine testing with such materials and are within the scope of this invention.

For electrochemical cell stacks, bipolar plates were formed with flow channels
25 defined by corrugated sheets 14 on both sides of electrically conductive flat sheet 16, as shown in Figure 2. Conductive sheets can be formed from any of a number of materials used in electrochemical cells, such as carbon, stainless steels, and the like. It will be understood that some applications may require a corrugated flow field adjacent only one electrode such that the bipolar plate will have only a single
30 corrugated sheet 14 that contacts one side of conductive sheet 16. For end cells,

conductive sheet 16 may terminate the stack; for other cells, a more conventional flow field may be provided on the second side of conductive sheet 14.

Figures 3 and 4 generically depict an electrochemical cell assembly and a cell stack, and illustrate the context for utilizing the benefits of the present invention.

5 Figure 3 is an exploded view, in cross-section, of an electrochemical cell assembly. Membrane electrode assembly 30 is formed from a proton conducting membrane 24, which is preferably a polymer electrolyte, that is placed between anode 22 and cathode 26 conductors, which are typically a conventional carbon cloth material. The membrane electrode assembly is formed with a catalyst that is selected for the
10 particular application. Suitable membrane materials and conductors may be selected in accordance with conventional teachings for gas reaction fuel cells or with teachings of U.S. Patent Application "Enhanced Methanol Utilization in Direct Methanol Fuel Cells", docket S-91,744, filed concurrently herewith by the named inventors herein, incorporated herein by reference.

15 Membrane electrode assembly 30 is placed between conductive corrugated sheets 14, which supply various fluids across the face of electrodes 22 and 26 through flow field passages defined by the corrugations. While Figures 3 and 4 illustrate bipolar plates 18 adjacent both electrodes 22, 26, it will be understood that the perforated flow field may be provided on only one electrode depending on the nature
20 of the reactant flow streams.

Perforations, e.g., perforations 12 in corrugated sheet 14 (Figures 1A and 1B), permit fluid interchange and mixing between flow channels in order to evenly distribute fluid flow over the surface of the electrodes, e.g., electrodes 22, 26. An even distribution of fluid flow enhances an even humidification of membrane 24 and the
25 removal of reaction water from along the cathode surface to enhance uniform reactant gas access to the electrode and membrane surface. Perforations 12 also permit fluids within the flow channels to contact the surfaces of electrodes 22, 24 over a larger surface area for increased reactant utilization and removal of reaction products.

In many stack designs, cooling plates are periodically interspersed between the
30 electrochemical cell assemblies. Instead of a reactant, coolant is distributed in some

manner across the plate. Since the cooling plates basically need to satisfy the same requirements as the bipolar plates (e.g., conductivity, strength, flow distribution, etc.) and may be configured such that one side distributes coolant and the other side a reactant, etc., such components will also be considered under the general term

5 “bipolar plates”.

In Figure 4, a plurality of membrane electrode assemblies 30 are placed alternately in series with bipolar plates 18, which serve to electrically connect in series anodes 22 and cathodes 26 of adjacent cells (see Figure 3 for electrochemical cell assembly references) to form electrochemical cell stack 40. End plates 32 and 34
10 contact end ones of corrugated flow devices 14 and are loaded to compress the stack of bipolar plates 18 and membrane electrode assemblies 30 between end plates 32 and 34

Both single cell and stacked fuel cell configurations in accordance with Figures 3 and 4 have been constructed and tested. As discussed below, a five-cell fuel cell
15 stack with liquid anode feed (1 M methanol (MeOH)) showed very good performance, even with low stoichiometric ambient air flow (3x oxygen consumption rate) at the test elevation (0.76 atm). The performance was achieved with a very low pressure drop of 1 in. of water across the stack and with a compact packaging of 2 mm per cell. These results signify an enhanced power density with low parasitic power losses from
20 pressure drops enabled by the flow field device of the present invention.

Test fuel cells were formed from membrane/electrode assemblies prepared using a 50 cm² Nafion® 117 membrane catalyzed with PtRu at 8 mg/cm² on the anode side and with Pt at 6 mg/cm² on the cathode side. The anode feed was 1 M MeOH at 2 cc/min and the cathode feed was 0.76 atm air at 314 standard cc/min. Initial tests
25 showed that performance was substantially reproducible between the five cells and was relatively constant down to air stoichiometry as low as two. In an extended test up to 2000 hours, the stack demonstrated a high efficiency of air distribution along the active cathode area by the corrugated/perforated configuration in spite of a high liquid water content at the cathode.

Several operating factors that might affect the stack performance were investigated. With methanol flow channels in a vertical orientation, the individual cells in the stack performed more uniformly than when the methanol flow channels were in a horizontal orientation. This might be due to the upward movement of CO₂ along vertically oriented flow channels to assist methanol distribution over the active area. But the difference in performance between vertical and horizontal channel orientation was relatively small, e.g., about 0.02 volts, or about 5%, at 150 mA/cm². In a preferred configuration, the liquid flow channels may be oriented in a horizontal direction and the liquid/gas flow channels in a vertical direction.

A chlor-alkali electrochemical cell was constructed using the bipolar plate configuration of the present invention in an "oxygen depolarized cathode" configuration. The oxygen electrode was an E-TEK commercial "ELAT" type electrode with, e.g., 80% Pt/C catalyst at a loading of 5 mgPt/cm². The cathode catalyst layer was part of this carbon cloth electrode structure, and the electrode was pressed mechanically into the (carboxylate) surface of a bilayer, sulfonate/carboxylate membrane.

The electrolysis cell was thus operated as a two-compartment cell, i.e., with no gap between the oxygen electrode and the membrane. This implies that the oxygen supply to the cathode catalyst and caustic removal from the catalyst have to take place through the same backing layer of the cathode. In three-compartment configurations of the oxygen depolarized cell structures, the gas feed and the collection of caustic reaction products are separated. The two-compartment configuration has advantages of simplicity and elimination of a liquid filled compartment from the cathode structure, but places special demands on the efficiency of the cathode flow field, which must effectively remove liquid caustic products from the cathode backing.

In accordance with one aspect of the present invention, the chlor-alkali cell had perforated/corrugated bipolar plates as described above, with the 316SS material gold plated for corrosion protection for the experimental. A silver plating or other corrosion resistant, electrically conductive coating can be used in a production environment.

The corrugations were positioned vertically in the cell. Manifolding of the oxygen feed

to the corrugated/perforated flow field was implemented with two simple inlet and outlet horizontal grooves that were machined in a graphite block serving as a current collector behind the metal flow field. A silver coated nickel plate or other suitable coated material can replace the graphite block in a more commercial configuration.

- 5 Cell test results are shown in Table A and show very similar cell voltages to those obtained with a machined serpentine flow field in a graphite block. A cell voltage of 1.9V-2.0V was measured for both flow fields at a current density of 0.4 A/cm². At a higher current density of 0.6 A/cm², the initial cell voltage was similar

	Current Density (A/cm ²)		Cell Voltage (V)		Voltage Drift (mV/hr)	
	1	2	1	2	1	2
Serpentine Flow Field	0.4	0.6	1.9-2.0	2.1-2.3	0.2-0.3	0.3-0.5
Perforated Flow Field	0.4	0.6	1.9-2.0	2.25	0.1 mV/day	stable

- 10 to that obtained using machined graphite cathode hardware (2.25 V), but the performance stability obtained with the perforated corrugated flow field was significantly superior. The voltage drift was only around 0.1 mV/day instead of 0.1 mV/hour. The cell operated continuously at 0.6 A/cm², practically maintaining its initial
- 15 performance. It is believed this stability arises from the improved ability of the flow field of the present invention to remove liquid products from the cathode. Thus, the flow field of the present invention provides good, highly stable cell performance, and has a significant cost advantage over other chlor-alkali cell hardware.

- 20 The foregoing description of the invention has been presented for purposes of illustration and description and is not intended to be exhaustive or to limit the invention to the precise form disclosed, and obviously many modifications and variations are possible in light of the above teaching. The embodiments were chosen and described in order to best explain the principles of the invention and its practical application to

thereby enable others skilled in the art to best utilize the invention in various embodiments and with various modifications as are suited to the particular use contemplated. It is intended that the scope of the invention be defined by the claims appended hereto.

WHAT IS CLAIMED IS:

1. A bipolar plate assembly for use in an electrochemical cell comprising:
a smooth electrically conductive sheet having first and second sides; and
a first corrugated sheet of conductive material in electrical contact with the first
side of the conductive sheet, having corrugations to define flow channels on the
corrugated sheet, and having perforations in the material to permit fluid transfer
5 between alternating flow channels.
2. A bipolar plate assembly according to Claim 1, further including:
a second corrugated sheet of conductive material in electrical contact with the
second side of the conductive sheet, having flow channels on the corrugated sheet,
and having perforations in the material to permit fluid transfer between alternating flow
5 channels.
3. A bipolar plate assembly according to Claim 2, where the second
corrugated sheet has a flow channel alignment that is perpendicular to a flow channel
alignment of the first corrugated sheet.
4. A bipolar plate assembly according to Claim 1, where the perforations in
adjacent rows are offset from one another.
5. A bipolar plate assembly according to Claim 2, where the perforations in
adjacent rows are offset from one another.
6. An electrochemical cell comprising:
a membrane electrode assembly having an anode and a cathode;
a bipolar plate in electrical contact with the cathode, comprising:
a smooth electrically conductive sheet having first and second sides; and
5 a first corrugated sheet of conductive material in electrical contact with
the conductive sheet, having corrugations to define flow channels on opposite sides of
the corrugated sheet, and having perforations in the material to permit fluid transfer
between the alternating flow channels.
7. A bipolar plate assembly according to Claim 6, further including:
10 a second corrugated sheet of conductive material in electrical contact with the
second side of the conductive sheet, having flow channels on the corrugated sheet,

and having perforations in the material to permit fluid transfer between alternating flow channels.

8. An electrochemical cell according to Claim 7, where the second corrugated sheet has a flow channel alignment that is perpendicular to a flow channel alignment of the first corrugated sheet.

9. An electrochemical cell according to Claim 6, where the perforations are arranged in rows that further permit fluid transfer to and from an adjacent anode or cathode.

10. An electrochemical cell according to Claim 7, where the perforations are arranged in rows that further permit fluid transfer to and from an adjacent anode or cathode.

11. An electrochemical cell according to Claim 9, wherein the perforations in adjacent rows are offset from one another.

12. An electrochemical cell according to Claim 10, wherein the perforations in adjacent rows are offset from one another.

13. An electrochemical cell stack comprising:
a plurality of electrochemical cells mechanically and electrically connected in series where each one of the cells comprises:

a membrane electrode assembly having an anode and a cathode;

5 a bipolar plate in electrical contact with the cathode, comprising:

a smooth electrically conductive sheet having first and second sides; and

a first corrugated sheet of conductive material in electrical contact with the conductive sheet, having corrugations to define flow channels on opposite sides of the corrugated sheet, and having perforations in the material to permit fluid transfer
10 between the alternating flow channels.

14. A bipolar plate assembly according to Claim 13, further including:

a second corrugated sheet of conductive material in electrical contact with the second side of the conductive sheet, having flow channels on the corrugated sheet, and having perforations in the material to permit fluid transfer between alternating flow
15 channels.

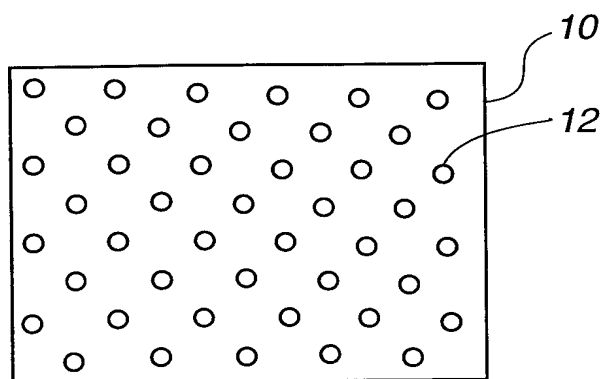
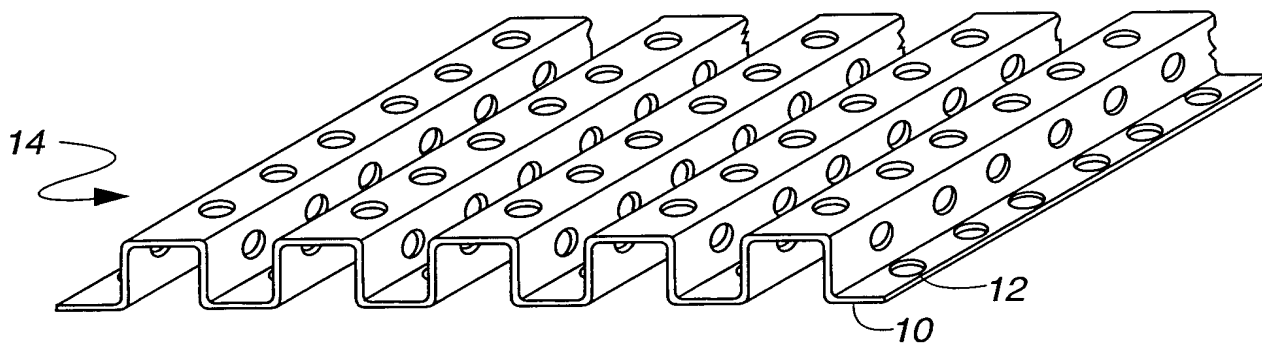
15. An electrochemical cell stack according to Claim 14, where the second corrugated sheet has a flow channel alignment that is perpendicular to a flow channel alignment of the first corrugated sheet.

16. An electrochemical cell stack according to Claim 13, where the perforations are arranged in rows that further permit fluid transfer to and from an adjacent anode or cathode.

17. An electrochemical cell stack according to Claim 14, where the perforations are arranged in rows that further permit fluid transfer to and from an adjacent anode or cathode.

18. An electrochemical cell stack according to Claim 16, wherein the perforations in adjacent rows are offset from one another.

19. An electrochemical cell according to Claim 17, wherein the perforations in adjacent rows are offset from one another.

**Fig. 1A****Fig. 1B**

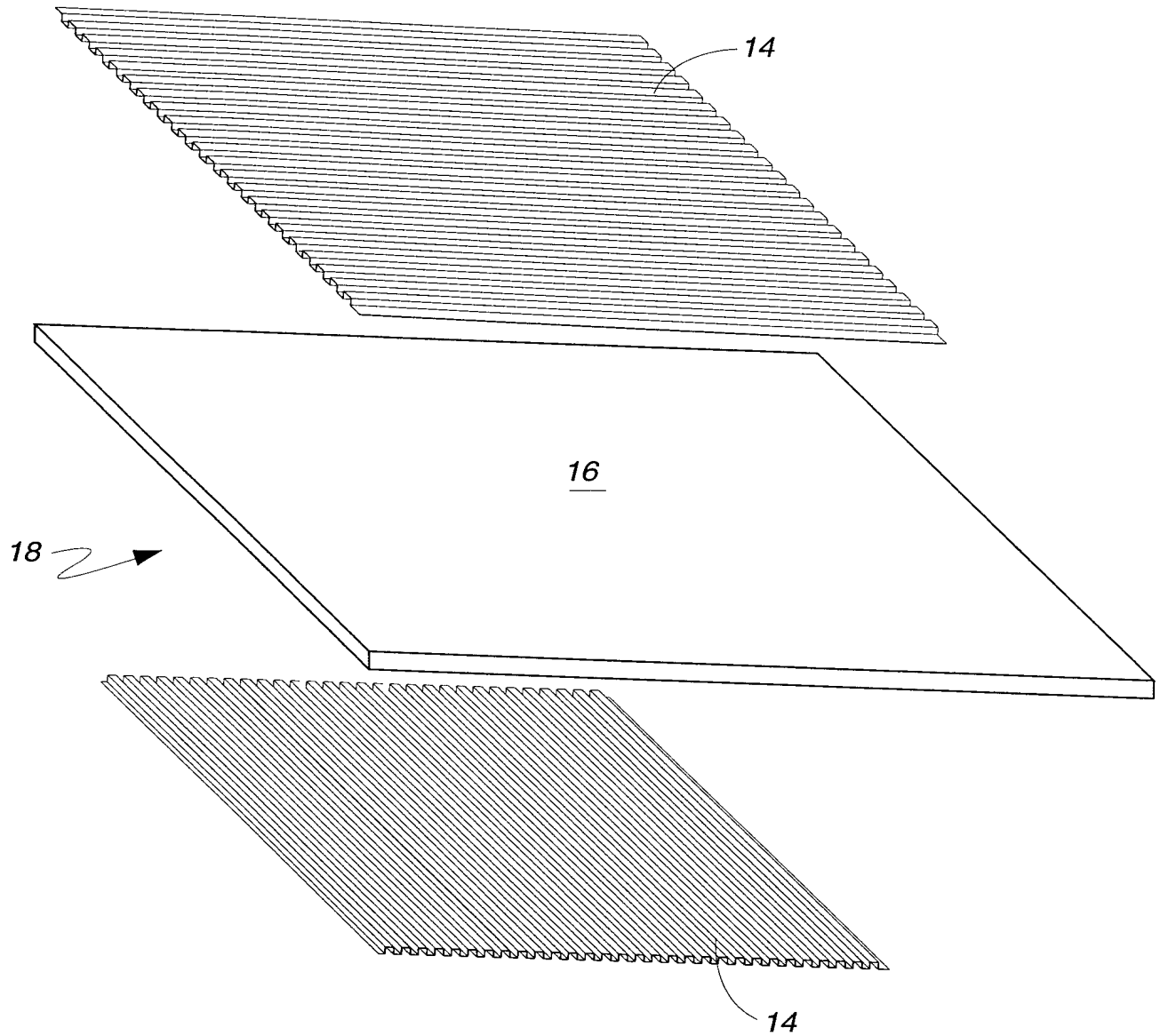
**Fig. 2**

Fig. 3

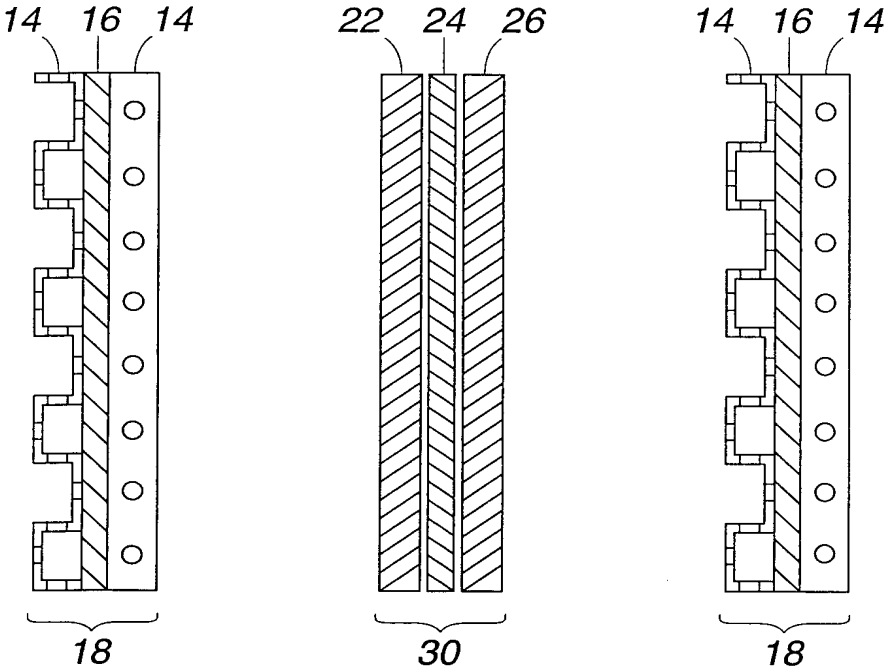
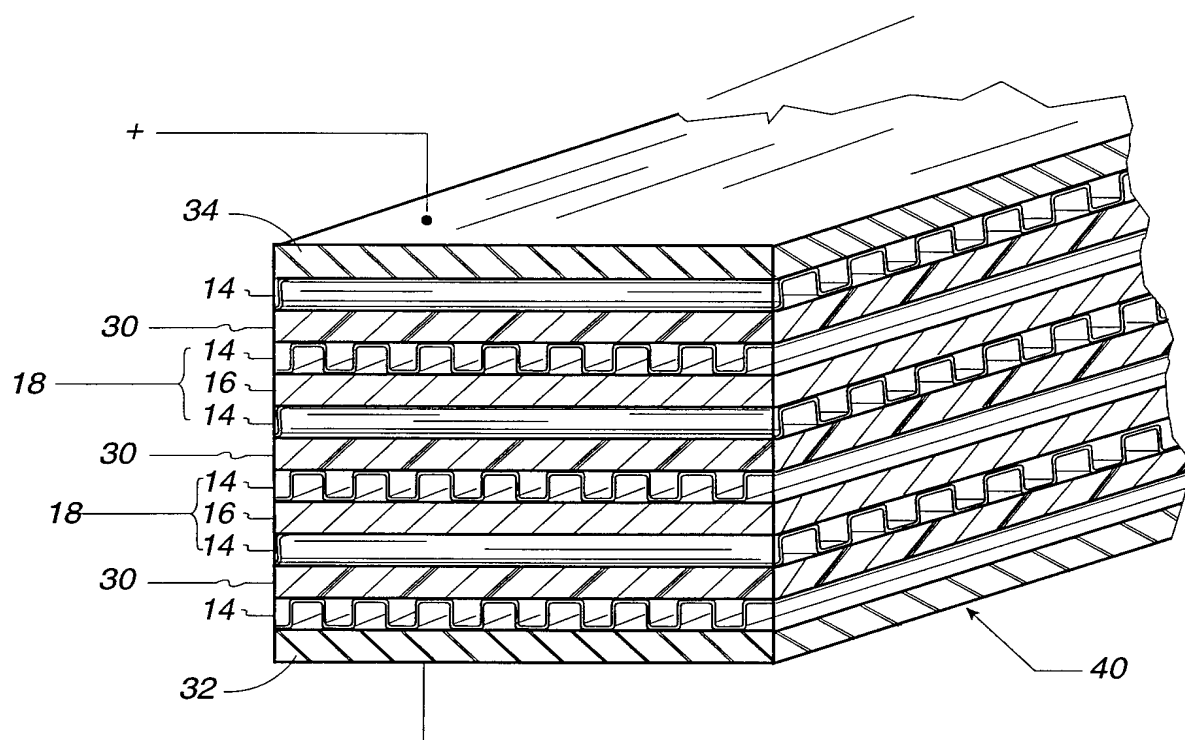


Fig. 4

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US00/32542

A. CLASSIFICATION OF SUBJECT MATTER

IPC(7) : H01M 8/04, 8/24

US CL : 429/32, 34

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 429/26, 30, 32, 34, 210

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EAST, WEST

search terms: fuel adj cell, corrugat\$, perforat\$

(\$ is a truncation symbol)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 3,432,357 A (DANKESE) 11 March 1969, col. 11, lines 27-29, col. 13, lines 6-9 and 35-38, figs. 1, 2, and 38.	1-19
X	US 4,788,110 A (BERNARD) 29 November 1988, col. 3, lines 19-45, figs. 1 and 2.	1
X	US 5,563,003 A (SUZUKI et al) 08 October 1996, col. 3, lines 33-44, figs. 1 and 3.	1-3
X	US 5,776,624 A (NEUTZLER) 07 July 1998, col. 1, line 66-col. 2, line 5, fig. 6.	1, 6, 13
A, P	US 6,117,580 A (NITSCHKÉ et al) 12 September 2000, col. 1, lines 59-65.	1-19
A	JP 63-086,361 A (HITACHI) 16 April 1988, figs. 1, 2 and 7-9.	1-19

☒ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A" document defining the general state of the art which is not considered to be of particular relevance	"X" document of particular relevance, the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"E" earlier document published on or after the international filing date	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"&" document member of the same patent family
"O" document referring to an oral disclosure, use, exhibition or other means	
"P" document published prior to the international filing date but later than the priority date claimed	

Date of the actual completion of the international search

07 MARCH 2001

Date of mailing of the international search report

10 APR 2001

Name and mailing address of the ISA/US
Commissioner of Patents and Trademarks
Box PCT
Washington, D.C. 20231

Facsimile No. (703) 305-3230

Authorized officer

STEPHEN J. KALAFUT

Telephone No. (703) 308-0661

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US00/32542

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	JP 5-029,009 A (HADO et al) 05 February 1993, fig. 1.	1-19
A	FR 2,067,158 A (RHONE-POULENC) 17 November 1969, page 3, lines 24-28, fig. 1.	1-19